Global Warming Potentials

Discussion Article

Characterisation Factors for Greenhouse Gases at a Midpoint Level Including Indirect Effects Based on Calculations with the IMAGE Model

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Abstract

Background, Aims and Scope. The traditional method of using Global Warming Potentials (GWPs) to assess the effects of climate change in Life Cycle Assessment (LCA) does not account for indirect atmospheric effects like saturation effects and emissions of ozone precursors. The aim of this paper is to provide GWP values for LCA purposes of the most important climate related gases including indirect effects and assess whether these values are dependent of different background conditions and size of change in emissions fluxes.

Methods. In order to reflect atmospheric interactions between direct and indirect greenhouse gases, an atmospheric climate and chemistry model needs to be used to determine GWP values. Here, the IMAGE model is used to determine GWP values for a time horizon of 100 years. Different runs were performed to test the outcomes against various background emission scenarios and various sizes in emission fluxes.

Results and Discussion. The GWP values for CO_2, N_2O and CH_4 depend on the chosen scenario and are lower under scenarios with higher background emissions. The GWPs of Halons, PFCs and SF_6 are scenario and emission flux independent. The GWPs of HFCs increase with higher background emissions and higher additional emission fluxes, whereas GWPs of CFC are scenario independent, but change when other emission fluxes are applied. Finally, GWPs of HCFCs are higher in scenarios with higher background emission scenarios, and decrease when larger emission fluxes are applied. The GWP values calculated with IMAGE with direct effects only are comparable with IPCC values. Inclusion of the indirect atmospheric effects changes some of the values positively or negatively. For CFCs this results in a value up to 70% lower. For Halons this results in a value up to 1000% lower, resulting in a strong negative GWP. Inclusion of indirect effects increases the GWP of CH_4 by 50%. Newly introduced greenhouse gases which have only indirect effects are given here a GWP. SO_2 has a negative GWP which depends on both the flux size and the chosen background scenario. CO and NMVOC have a positive flux-and-scenario dependent GWP. The GWP of NO_x can, dependent on de chosen conditions, be positive or negative.

Conclusions. The GWP values of this paper are a first attempt to provide a consistent set of GWP values for all direct and indirect greenhouse gases, including differences in GWP values per background scenario and flux size. The inclusion of indirect effects in GWP values causes large differences in some important greenhouse gases, which should not be ignored in LCA-analyses. We suggest for LCA purposes to use GWPs including indirect effects for marginal change and for the most realistic background scenario. However, existing uncertainties in the indirect effects of greenhouse gases demand for a better understanding of the importance of these effects.

Keywords: Climate change; global warming potentials (GWP); IMAGE; impact assessment; LCA; midpoint characterization factors

Introduction

It is the purpose of this commentary to discuss the sensitivity of Global Warming Potentials (GWP) of the most important climate-related gases towards the inclusion of indirect climate effects. We welcome critical comments to our proposal.

Climate change is one of the impacts considered in Life Cycle Assessment (LCA) (Udo de Haes et al. 1999a,b, Jolliet et al. 2004). The rapid increase of atmospheric greenhouse gases concentrations due to energy, industrial and land use emissions causes significant changes in the climate. Climate change affects ecosystems and possibly a wide range of human health aspects (IPCC 2001a). The Global Warming Potential (GWP) is generally used to indicate the impact of climate change in LCA (Guinée 2001). This indicator gives information about the relative impact of greenhouse gases; it is a measure of the total increase in radiative forcing per unit of emission. GWP is a useful measure to compare and sum the effects of different greenhouse gases. Disadvantages include not accounting for geographical differences (Eickhout et al. 2003) and the arbitrary time horizon (Smith & Wigley 2000a,b, Van Vuuren et al. 2006). In comparison with alter-

native methods it is however the most feasible method for political purposes (Fuglestvedt 2003).

GWPs used for LCA (Guinée et al. 2001) are based on the list available from IPCC for direct greenhouse effect of the gases. Because of the great uncertainties about their magnitudes, indirect effects were excluded in deriving the IPCC GWPs (IPCC 2001, 2007). Indirect effects on the GWP arise from the formation and depletion of other atmospheric substances due to an emission of a gas. Effects of the so-called indirect greenhouse gases CO, NO_x, NMVOC and SO_2 were not considered in the IPCC-GWPs. These substances are no greenhouse gases themselves, but have an effect on the formation of tropospheric ozone, methane and sulphate aerosols, which do influence the radiative forcing (IPCC 2001b). In addition, well-known greenhouse gases have indirect effects as well. Chlorides such as chlorofluorcarbons (CFCs), and hydrochlorofluorcarbons (HCFCs) for instance, increase the radiative forcing, but are also related to the depletion of stratospheric ozone, causing a cooling of the atmosphere. This indirect effect can be even larger than the direct effects (Daniel et al. 1995, WMO 2003, Velders et al. 2005). For the purpose of LCA impact assessment, it may be worthwhile to consider the indirect effects, in spite of the great uncertainties involved.

Recent studies in the LCA field focused on the adaptation of existing risk assessment models to derive and improve characterisation factors for various impact categories, such as acidification (Seppälä et al. 2006), photochemical ozone formation (Labouze et al. 2004), stratospheric ozone depletion (Hayashi et al. 2006), and toxicity (McKone & Hertwich 2001). In this study we use a similar approach for the improvement of characterisation factors for climate change. The Integrated Model for the Assessment of the Global Environment (IMAGE) simulates changes in climate variables and land use patterns as result of changes in demographic, technological and economic changes in society through changes in emissions of greenhouse gases (Alcamo 1994, IMAGE Team 2001). The model can consistently calculate the increase in radiative forcing as a result of emissions of well-known greenhouse gases (CO_2, N_2O, CH_4, Hydrofluorcarbons (HFCs), Perfluorcarbons (PFCs) and SF_6), priority air pollutants (SO_2, NO_x, CO and non-methane volatile organic compounds (NMVOC)) and ozone depleting pollutants (Halons and Chlorides). The IMAGE 2.2 model includes feedbacks from the ocean and the terrestrial environment to the atmospheric environment and vice versa (Alcamo 1994). The IMAGE model has been documented in detail (Alcamo 1994, Alcamo et al. 1998, IMAGE Team 2001, MNP 2006) and discussed in scientific papers (e.g. Leemans & Eickhout 2004, Strengers et al, 2004, Eickhout et al. 2006, Van Vuuren et al. 2007). Moreover, the IMAGE model has been used in several global assessments to address issues like greenhouse gas emissions (IPCC 2000), global environmental change (GEO3 2002) and ecosystem goods and services (MA 2005). The latest scientific review of the IMAGE model concluded the general appreciation in policy circles has been built on the scientific credibility of the IMAGE model (Hordijk et al. 2007). In the latest IPCC Fourth Assessment Report, results of the IMAGE model are also used for conclusions on climate mitigation (IPCC 2007).

This paper focuses on the atmospheric effect of greenhouse gases only. In IMAGE 2.2 the influence of four emission scenarios are assessed, reflecting different views on how the world may develop in the next 100 years. In the Special Report on Emission Scenarios (SRES) (IPCC, 2000) these so-called SRES scenarios are described. The goal of this paper is (i) to calculate GWPs of direct and indirect greenhouse gases including indirect atmospheric effects with the IMAGE 2.2 atmospheric climate and chemistry model (Eickhout et al. 2004) for use in Life Cycle Impact Assessment (LCIA) and (ii) test the linearity of these IMAGE-based midpoint characterization factors against the chosen emission change applied and the chosen emission background scenario.

1 Methodology

1.1 IMAGE

1.1.1 Model structure

IMAGE 2.2 describes the climate change systems in all its complexity (Alcamo 1994). Although still a simplification, it includes many feedbacks to show the influence of most aspects of climate change. **Fig. 1** shows a schematic overview of the IMAGE model.

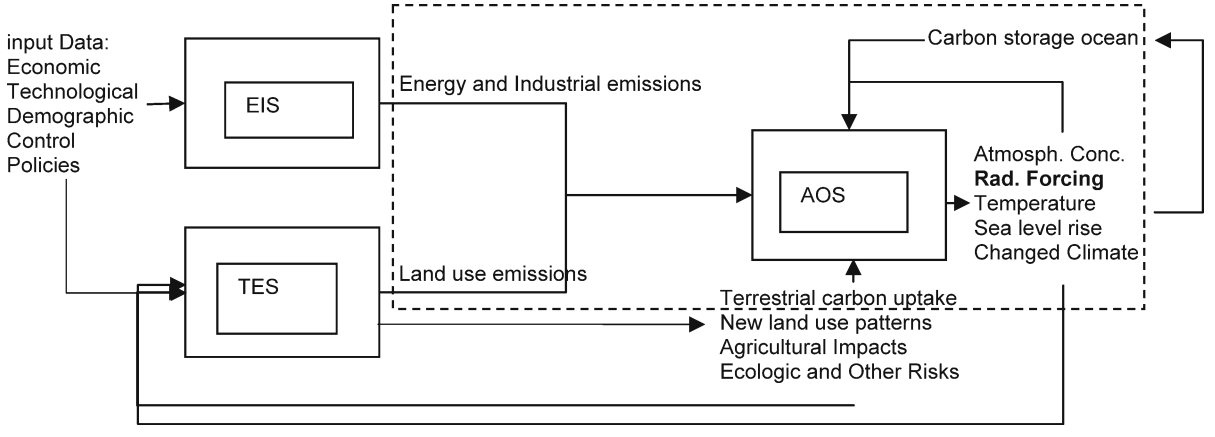

Fig.1: Schematic overview of the interrelations of the three systems in the IMAGE model. Within the dotted line, the part of the model used for the study in this paper is selected

The model consists of three systems:

- The Energy Industry System (EIS) computes all emissions of greenhouse and air polluting gases out of energy use and industrial activities with economic and population driving forces as main input of the system
- The Terrestrial Environment System (TES) calculates food demand, land-use changes and changes in carbon uptake capacity, resulting in land use emissions of greenhouse and air polluting gases; it includes feedbacks of climatic change on the terrestrial environment, like CO_2 fertilization and the temperature effect on soil respiration
- The Atmosphere Ocean System (AOS) computes radiative forcing, temperature change and sea-level rise due to changes in atmospheric concentrations of greenhouse gases as a result of the emission of gases out of the two previous systems

1.1.2 Climate feedback

Feedback from climate change also influences the system. The terrestrial uptake of carbon, calculated in the TES, is influenced by climate change. Furthermore, temperature change is used to calculate oceanic uptake of CO_2. In this study we are interested in the effect of small additional emissions on radiative forcing. To get a clear view on how the individual substances behave atmospherically, the feedbacks of temperature on the terrestrial and oceanic system are left out of the calculation. Land use emissions and terrestrial carbon uptake are only considered exogenously (dependent per scenario) and oceanic carbon uptake is only dependent of the atmospheric CO_2 concentration determined by AOS. In Fig. 1, the parts of the model which are used are shown within the dotted line. More detail on how these processes are implemented in IMAGE is given in Eickhout et al. (2004).

1.1.3 Calculation of concentration of greenhouse gases

The calculation of the concentration of a greenhouse gas from the emissions is given by the equation:

$$\frac{\Delta[GHG]}{\Delta t} = cv_{GHG} \cdot E_{GHG} - k_{GHG}[GHG] \quad (1)$$

where [GHG] is the atmospheric concentration of greenhouse gas GHG (in ppmv), E_{GHG} the emission of the GHG in kg/yr, $k_{GHG\,the}$ reaction rate which is the reciprocal of the atmospheric lifetime (yr^{-1}) and cv_{GHG} a mass-to concentration conversion factor (ppmv/kg). The lifetimes of N_2O, CFCs, Halons and PFCs and SF_6 are assumed to be constant.

The lifetimes of the gases CH_4, HCFCs and the HFCs are related to the OH abundance:

$$k_{GHG} = k_{GHG+OH} \cdot [OH] + k_{srat+bio} \quad (2)$$

where k_{GHG+OH} represents the oxidation of the GHG with OH, [OH] the concentration of OH-radicals and $k_{strat+bio}$ the loss rate to stratosphere and biosphere, which are assumed to be constant. A higher background concentration of OH radicals results in a lower lifetime of the chemical. This means that the chemical compound disappears faster out of the atmosphere.

Calculation of concentration of CO_2 is more complex given the uptake of carbon by oceans and terrestrial biosphere.

$$\frac{\Delta[CO_2]}{\Delta t} = cv_{GHG} \cdot \left(E_{CO_2} - S_{CO_2}\right) \quad (3)$$

The sinks (S_{CO2} in kg/yr) are the CO_2 uptake by vegetation, calculated in the TES, and the CO_2 uptake by the oceans, calculated in AOS. Here, sink fluxes from TES are pre-calculated and used as input exogenously and are not part of the simulation itself. The values of lifetimes, mass-to concentration factors and reaction rates are given in the supporting information.

1.1.4 Calculation of concentration of tropospheric ozone and OH-radicals

The indirect greenhouse gases CH_4, NO_x, CO and NMVOC form tropospheric ozone and OH-radicals.

$$ln[OH] = P_{CH_4} \cdot ln\,[CH_4] + P_{NO_x} \cdot E_{NO_x} \cdot P_{CO} \cdot E_{CO} \cdot P_{NMVOC} \cdot E_{NMVOC} \quad (4)$$

$$n[trop.O_3] = R_{CH_4} \cdot ln\,[CH_4] + R_{NO_x} \cdot E_{NO_x} \cdot R_{CO} \cdot E_{CO} \cdot R_{NMVOC} \cdot E_{NMVOC} \quad (5)$$

P and R in these formulas are empirically derived sensitivity coefficients to determine the change in OH abundance and tropospheric ozone concentration (IPCC, 2001b). The values are given in the supporting information. Mind that the values of P are, except for NO_x, negative while the values of R are positive. This means that with increasing concentrations of CH_4, CO and NMVOC the OH abundance decreases while the tropospheric ozone concentration increases. Natural emissions from sources like natural vegetation, volcanoes and oceans are included, although these are kept constant over time. Tropospheric ozone is a greenhouse gas. OH-radicals change lifetimes and thereby the concentration of the gases CH_4, HCFCs and the HFCs according to Eq. 2.

1.1.5 Calculation of change in radiative forcing

The radiative forcing due to halocarbons, tropospheric ozone and SF_6 is modelled as proportional to the calculated concentration (ppmv). The proportionality constant is the empirically derived radiative efficiency coefficient re_{GHG} ($Wm^{-2}ppbv^{-1}$). The gas-specific re_{GHG} values used are given in the supporting information.

$$RF_{GHG,t} = re_{GHG} \cdot [GHG]_t \quad (6)$$

The radiative forcing of CO_2 depends on pre-industrial concentrations:

$$RF_{CO_2} = re_{CO_2} \cdot ln\left(\frac{[CO_2]_t}{[CO_2]_{t0}}\right) \quad (7)$$

The radiative forcing due to CH_4 and N_2O is besides the pre-industrial concentration of the substances themselves also dependent on the pre-industrial concentration of each other. The equation of the RF of both substances is given as:

$$RF_{GCG} = re_{GHG}\sqrt{[GHG]_t} - \sqrt{[GHG]_{t0}} - f([GHG]_t, [GHG]_{t0}, [GHG2]_{t0}) \quad (8)$$

GHG is the greenhouse gas CH_4 or N_2O and GHG2 is the other greenhouse gas; in the equation of CH_4 this is N_2O and in the equation of N_2O this is CH_4. The function in Eq. 8 is given in the supporting information. Also the values of the pre-industrial concentrations of CO_2, CH_4 and N_2O are given there.

The negative radiative forcing due to ozone depletion of chlorinated chlorides and bromated halons in the stratosphere is represented by:

$$RF_{strat.O_3,t} = -0.001 \cdot \left[d_{Cl} \sum_i \left\{ NCl_i \cdot [GHG]_{i,t} \right\}^{1.7} + d_{Br} \sum_i \left(NBr_i \cdot [GHG]_{i,t} \right) \right] \quad (9)$$

where GHG is the concentration of the concentration of the chlorinated or bromated GHG and NCl_i and NBr_i are respectively the number of reactive chlorine or bromine atoms in GHG i. d_{Cl} and d_{Br} are the depletion coefficients of chlorine and bromine. These numbers are given in the supporting information.

The radiative forcing due to stratospheric water vapour is positively affected by a higher concentration of CH_4:

$$RF_{strat.H_2O,t} = 0.05 \cdot re_{CH_4} \cdot \left(\sqrt{[CH_4]_t} - \sqrt{[CH_4]_{t0}}\right) \quad (10)$$

Another forcing effect is the direct radiative forcing of tropospheric sulphate aerosols (Myhre 1998). This is a result of the emission of the indirect greenhouse gas SO_2.

$$RF_{aero-direct,t} = \frac{E_{SO_2,t}}{E_{SO_2,1990}} \cdot RF_{aero-direct,1990} \quad (11)$$

Sulphate aerosols cause also indirect effects. Indirect effects of tropospheric aerosols include the changing of the cloud albedo and amount due to cloud condensation and ice nuclei. For this effect also the natural stratospheric emission of SO_2 $E_{SO2\text{-}nat}$ is considered:

$$RF_{aero-indirect,t} = \frac{\log(1 + E_{SO_2,t} / E_{SO_2-nat})}{\log(1 + E_{SO_2,1990} / E_{SO_2-nat})} \cdot RF_{aero-indirect,1990} \quad (12)$$

The initial emission of SO2 in 1990 assumed is 71.6 Tg S. The initial radiative forcing in 1990 due to sulphate aerosols are –0.3 Wm-2 and –0.8 Wm-2 for direct and indirect effects respectively. The natural emission of SO_2 is assumed to be a constant amount of 34.4 Tg S.

1.2 Global warming potentials

IPCC (2001b) distinguishes two different measures for global warming: the Absolute Global Warming Potential (AGWP) and the Global Warming Potential (GWP). We define the AGWP as the increase over time in radiative forcing per kilogram emission change of a substance over a given time horizon:

$$\mathrm{AGWP(x)} = \frac{\Delta \mathrm{RF_{TH}}}{\mathrm{E_x}} \quad (13)$$

The nominator represents the change in radiative forcing at time TH. E_x is the yearly added emission flux. The AGWP is expressed in W m^{-2} kg^{-1} yr. The Global Warming Potential is the AGWP of a substance related to the AGWP of a reference gas, CO_2 (CO_2-eq.):

$$\mathrm{GWP(x)} = \frac{\mathrm{AGWP_{GHG}}}{\mathrm{AGWP_{CO_2}}} \quad (14)$$

The approach used by IPCC (2001b), is a so-called pulse-based method. This means that at time zero an emission pulse of a substance is released, which results in an instantaneous increase in the concentration in the atmosphere, whereafter the concentration gradually decreases over time due to reactions in the atmosphere. The increase in concentration corresponds with an increase in radiative forcing. The approach used in this paper is the flux-based method. In this case, the extra emission is added yearly, resulting in an increase of the radiative forcing towards a new steady state situation. The situation is shown in **Fig. 2**. Fluxes and pulses in a linear system yield the same characterization factor (Heijungs 1995).

From the year 2000 an extra emission flux has been added to the standard scenario up to the year 2100. The time horizon of 100 years was chosen because it is common in climate change research, as it is the average atmospheric lifetime of CO_2 (IPCC, 2001b). GWPs were calculated for CO_2, N_2O, CH_4, NO_x, SO_2, CO, NMVOC, the halocarbons (CFCs, HCFCs, HFCs and Halons), the PFCs and SF_6.

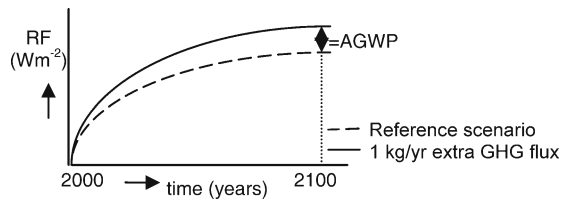

Fig. 2: Change in RF with the flux-based method

1.3 Simulations

In the evaluation of GWPs with IMAGE several choices were required. To assess the influence of the percentage of marginal change and the choice for a background emission scenario, the following simulations were performed.

1.3.1 Marginal emission change

We tested the linearity of the effect of change of emissions on the change in radiative forcing. The simulations were executed for different amounts of ΔE. We tested this linearity with respectively 1%, 10% and 100% extra emission of the total anthropogenic emissions in the year 2000. In a linear situation the RF will increase linearly with the emission what will lead to a GWP value which is the same for all the three emission changes. If this is not the case the GWP values for the three emission changes will be used to calculate the GWP that reflects a marginal emission change. This is done by making an exponential fit, shown in **Fig. 3**. The marginal change is the change closest to zero.

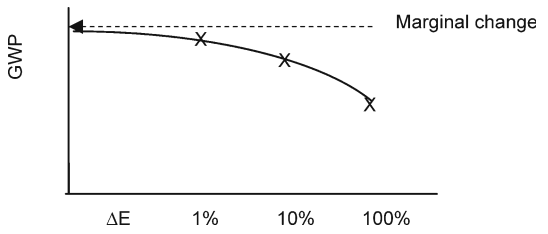

Fig. 3: The GWP for different scenario fluxes can be exponentially related. The GWP for the marginal emission change is the value for the smallest possible emission

1.3.2 Background emission scenarios

Marginal changes were made in the four different base scenarios presented in IMAGE 2.2. The Intergovernmental Panel on Climate Change (IPCC) uses these four so-called SRES scenarios A1, A2, B1 and B2 (IPCC, 2000). IPCC developed these multiple scenarios to explore the uncertainties behind potential trends in global developments and GHG emissions. The scenarios differ in how global regions interrelate, how new technologies diffuse, how regional economic activities evolve, how protection of local and regional environments is implemented, and how demographic structure changes (IPCC 2000). These different developments result in differences in projected emissions. These emissions are given in **Fig. 4**.

2 Results and Discussion

2.1 Direct GWPs

Table 1 and **2** show direct GWPs of greenhouse gases. 'Direct' here means that only the change in radiative forcing due to changes in the concentration of the substance itself is taken into account. Indirect effects, such as the change in radiative forcing due to changes in tropospheric ozone and lifetime changes of CH_4, HCFCs and HFCs and indirect forcing effects, are considered in the next result section. The reference value was the AGWP of 1% CO_2 change in the A1 scenario, being $9.06.10^{-14}$ $Wm^{-2}kg^{-1}yr$.

2.2 Sensitivity of direct GWPs to the chosen emission change

Table 2 shows the direct GWPs for the emission changes of 1%, 10% and 100% for the three main substances CO_2, CH_4 and N_2O. Larger emissions cause lower GWP values presenting a lower increase in radiative forcing per kilogram of added substance. Eq. 1 and 6, show that the change in

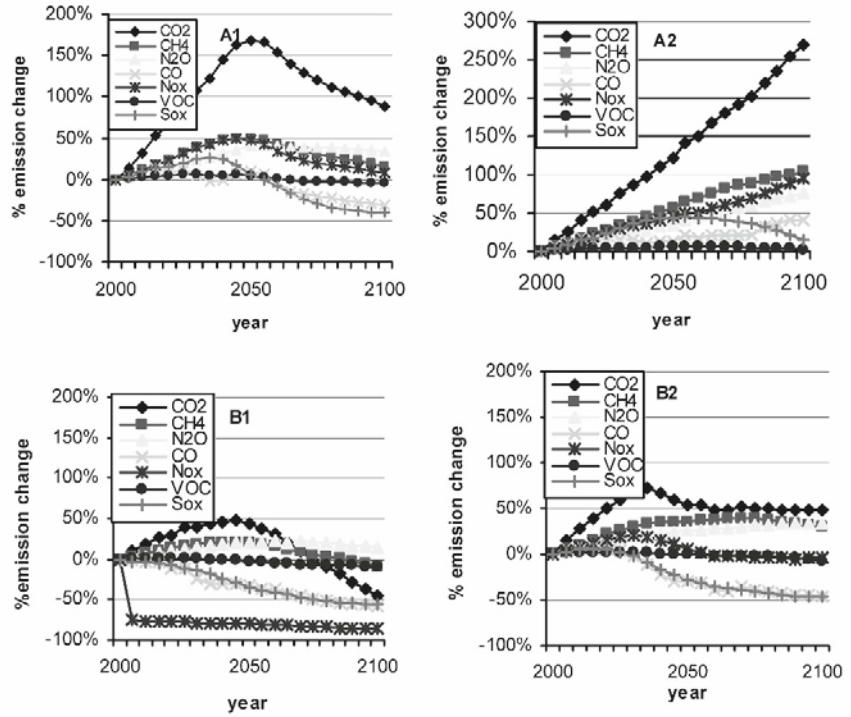

Fig. 4: The emission development in the 4 different scenarios. The emission development is given in relation to the year 2000

Table 1: Direct GWP values for the main greenhouse gases for different background scenarios and a 1% emission change relative to the total emission of the substance in the year 2000. Only direct effects of the substance on the radiative forcing are considered

Substance	A1	A2	B1	B2
CO_2	1	0.9	1.5	1.2
N_2O	276	268	288	281
CH_4	18	15	20	18
CFC11	4,800	4,800	4,800	4,800
CFC12	11,000	11,000	11,000	11,000
CFC113	6,200	6,200	6,200	6,200
CFC114	10,000	10,000	10,000	10,000
CFC115	7,500	7,500	7,500	7,500
CCl_4	1,800	1,800	1,800	1,800
$CH3CCl_3$	2,100	2,300	2,100	2,200
HCFC22	2,500	2,700	2,500	2,600
HCFC123	150	170	150	150
HCFC124	700	900[a]	800 [a]	800
HCFC141b	880	960	860	910
HCFC142b	3,000	3,300	3,000	3,100
HCFC225ca	700	800	700	800
Halon1211	1,300	1,300	1,300	1,300
Halon1301	7,200	7,200	7,200	7,200
CH_3Br	3	3	3	3
HFC23	13,100	13,200	13,100	13,100
HFC32	900 [a]	900	800	900
HFC43-10	5,800	5,600	5,000	5,200
HFC125	5,300	5,200	4,900	5,000
HFC134a	1,900	2,100	1,900	2,000
HFC143a	6,100 [a]	5,600	5,400	5,500
HFC227ea	4,900	5,300 [a]	5,000	5,200
HFC245ca	970	1,080	950	1,000
CF_4	6,100	6,100	6,100	6,100
C_2F_6	14,700	14,700	14,700	14,700
SF_6	23,400	23,400	23,400	23,400

[a] HCFC124, HFC32, HFC143 and HFC227 have relatively low emissions. For small emission changes of 1% it is possible that due to rounding errors the GWP values differ slightly from values calculated with larger fluxes

Table 2: The influence of different emission changes on the GWP of CO_2, N_2O and CH_4

Substance	1%	10%	100%
CO_2	1	0.98	0.82
CH_4	28	27	25
N_2O	276	275	265

concentration and radiative forcing due to concentration of most substances is related to a constant value, while for CO_2, CH_4 and N_2O (see Eq. 7 and 8) the relation between initial and actual concentration is involved. The lower GWP-values by larger contributions can be declared by the fact that the contribution to global warming is less per molecule when there are more molecules present in the atmosphere. This saturation effect happens as the forcing gradually saturates into absorption bands while the strongest absorption bands have already been taken (Eickhout et al. 2004). This effect was also found by Caldeira & Kasting (1993).

2.3 Sensitivity of direct GWPs for the chosen background scenario

Table 1 shows that background emissions play a role in the effect of contributions of CO_2, CH_4, N_2O, HCFCs, CH_3CCl_3 and HFCs irrespective of indirect influences. The scenarios with the highest background emissions (see Fig. 4), A2 and A1, give the lowest GWP values for CO_2, CH_4 and N_2O. This is the same type of result as seen from the assessment of different emission changes: higher background emissions of these substances results in a lower GWP per added kilogram of the substance. For HFCs, CH_3CCl_3 and HCFCs the scenarios with the high background concentrations result in higher values of the GWP. These gases do not have a constant lifetime but one that depends on the concentration of OH-radicals (see Eq. 2). The OH abundance decreases with higher emissions of CH_4, NO_x, CO and NMVOC (see Eq. 5). This causes a higher lifetime for the HCFCs, $CH3CCl_3$, CH_4, and the HFCs, which results in a higher concentration growth and therefore a higher RF (IPCC, 2001b). CFCs, Halons and PFCs (including SF_6) are independent of background scenarios.

2.4 Indirect GWP effects

Table 3 shows the GWPs of the greenhouse gases including indirect effects. The reference value was the same as the one used in Table 1 and 2 ($9.06.10^{-14}$ $Wm^{-2}kg^{-1}yr$). As can be seen in the table some substances have additional radiative forcing effects besides the direct effects shown in Table 1 and 2. Additional effects are seen for CH_4, CFCs, HCFCs and halons. The ozone precursors NO_x, CO, NMVOC and SO_2 did not have direct effects, but they do show indirect effects.

The values of the total GWPs are significantly lower for CFCs and the halons. This is a result of the depletion of stratospheric ozone, which leads to a negative contribution to the radiative forcing resulting in a net lower GWP. As can be seen by Eq. 9 this effect caused by chlorine atoms causes an exponential effect in the change in radiative forcing. The bromine atoms do not have this exponential effect. The GWP-values for CFCs for small emissions are about 30% lower then the ones including only direct effects. With additional emissions up to 100% (see Table 3) the GWP values can even be 70% lower. In the case of halons the indirect cooling effect is about 10 times higher then the direct warming effect, resulting in a negative GWP value. This results in a strong net negative effect of these substances. The indirect effect for HCFCs is generally low, about 1%. These outcomes were also found by Daniel et al (1995). Interestingly, the combination of effects for HCFCs leads to lower GWP values in scenarios with low background emissions due to more OH abundance (see Section 2.3), but higher GWP values with higher emission fluxes due to more stratospheric depletion.

Substances like CH_4, CO, NMVOC and NO_x have multiple indirect effects. They include the formation of the greenhouse gas tropospheric ozone and the OH radical. The last effect affects the lifetime of CH_4, HCFCs and HFCs. CH_4, CO, NMVOC to cause positive contributions through both effects, while NO_x has a positive effect through tropospheric ozone but a negative effect through OH abundance. This can result in either a net negative or a net positive GWP

Table 3: GWPs of with IMAGE 2.2 calculated greenhouse gases including direct and indirect effects (substances with only direct effects are left out of this table)

	A1			A2			B1			B2		
Substance	**1%**	**10%**	**100%**	**1%**	**10%**	**100%**	**1%**	**10%**	**100%**	**1%**	**10%**	**100%**
CH_4	28	27	25	21	20	19	31	31	27	26	26	24
CO	2.7	2.7	2.8	2.8	2.8	2.8	2.4	2.4	2.4	2.6	2.6	2.6
NO_x	–1.2	–1.1	–0.4	–1.3	–1.2	–0.9	2.5	2.6	3.1	0.0	0.1	0.5
NMVOC	8.2	8.2	8.2	8.2	8.2	8.2	7.2	7.3	7.3	7.9	7.9	7.9
SO_2	-94	–90	–71	–59	–58	–52	–118	–112	–82	–101	–97	–74
CFC11	3,200	3,000	1500	3,200	3,000	1,500	3,200	3,000	1,500	3,200	3,000	1,500
CFC12	6,000	5,700	2,700	6,000	5,700	2,700	6,000	5,700	2,700	6,000	5,700	2,700
CFC113	4,700	4,600	3,600	4,700	4,600	3,600	4,700	4,600	3,600	4,700	4,600	3,600
CFC114	9,800	9,700	9,700	9,800	9,700	9,700	9,800	9,700	9,700	9,800	9,700	9,700
CFC115	7,400	7,300	7,300	7,400	7,300	7,300	7,400	7,300	7,300	7,400	7,300	7,300
CCl_4	1,300	1,200	640	1,300	1,200	640	1,300	1,200	640	1,300	1,200	640
CH_3CCl_3	2,000	1,900	1,000	2,300	2,100	1,000	2,000	1,800	1,000	2,100	1,900	1,000
HCFC22	2,500	2,400	2,000	2,700	2,600	2,200	2,400	2,400	2,000	2,500	2,500	2,100
HCFC123	150	150	150	170	170	170	150	150	150	150	150	150
HCFC124	700	700	700	900 [a]	800	800	800 [a]	700	700	800	800	800
$HCFC141_b$	870	850	740	960	930	800	860	840	730	900	880	760
$HCFC142_b$	3,000	3,000	2,900	3,300	3,200	3,100	3,000	2,900	2,800	3,100	3,000	2,900
HCFC225	700	700	700	800	800	800	700	700	700	700	700	700
Halon1211	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000	–12,000
Halon1301	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000	–62,000
CH_3Br	–824	–824	–824	–824	–824	–824	–824	–824	–824	–824	–824	–824

[a] HCFC124, HFC143 and HFC227 have relatively low emissions. For small emission changes of 1% it is possible that due to rounding errors the GWP values slightly differ from values calculated with larger fluxes.

depending on the chosen background conditions. CH_4 causes, besides these effects, also a forcing by water vapour (see Eq. 10). SO_2 causes the formation of sulphate aerosols. These particles have direct and indirect forcing effects (see Eq. 11 and 12). Both effects cool the atmosphere. The greenhouse gases CO_2 N_2O, the HFCs and the PFCs are found to have no indirect contributions and are therefore left out in Table 3.

2.5 Direct and total GWPs in relation to previous research

Table 4 shows the GWPs of the gases of scenario A1 with their direct and their total effects from Table 1 and Table 3 for marginal emission changes. These values are compared with the values of IPCC (2001b). The table shows the GWPs for marginal contribution. For most greenhouse gases the GWP for 1, 10 and 100% emission change are approximately equal. As stated in section 2.2 CO_2, CH_4, N_2O, NMVOC, CO, NO_x and the chlorides are dependent on the chosen emission change. Generally the GWP for marginal change is approximately the same as the GWP for 1%. The marginal-change GWP of CFC11 and CFC12 is however smaller then the one for 1%. For these substances the exponential fit is used as given in section 1.3.1. IPCC uses a reference value based on the Bern carbon cycle model assuming a constant future concentration of CO_2. This value is $8.99\ 10^{-14}$ W m^{-2}kg (IPCC 1994, 2001b).

The direct GWPs calculated in this study are close to the values listed in the IPCC report (2001b). Noticeable differences can be seen for CH_4, chlorides, halons and HFCs. In the calculation of CH_4 IPCC did account for the indirect effects formation of stratospheric water vapour and tropospheric ozone. The effect of changing lifetimes was, however, not included. An increase in lifetime due to a lower OH concentration (see Eq. 1, 2 and 4) affects the GWP also positively. This makes that the IPCC-value for CH_4 lies in between the value of direct and total GWP. The calculated GWPs for NO_x CO, NMVOC and SO_2 were not listed before in the IPCC report. However, Fuglestvedt et al (1996) investigated the indirect effects of CH_4, CO and NO_x through changes in tropospheric ozone and CH_4 resulting in estimated GWP values of 30 for CH_4, 3 for CO and 'slightly negative' for NO_x from surface sources. These results compare well with the ones found in this study.

The results of the GWP values calculated with IMAGE including direct effects only are comparable to GWPs of IPCC (2001b). The total GWP values found in this study, which includes indirect effects of chlorides and halons, are much lower and compare well with results obtained by Daniel et al. (1995), WMO (2003) and Velders et al. (2005), who did similar studies. However, since these studies used different emission scenarios it is difficult to directly compare the results.

The GWP values for HCFCs and HFCs differ the most from the values of IPCC. This can be seen as the effect of the way OH radical reaction rates are implemented in the GWP calculations. In IMAGE the lifetime of these GHGs explicitly depends on the presence of OH-radicals. The concentration of this radical is variable and is influenced by the emissions

Table 4: GWPs of with IMAGE 2.2 calculated greenhouse gases including indirect effects. In the last column the values of the GWPs as published in the report of IPCC (2001b) are shown

	GWP_{100} direct	GWP_{100} total	IPCC, 2001
CO_2	1	1	1
N_2O	276	276	296
CH_4	18	28	23
CO	0	3	
NO_x	0	–1	
NMVOC	0	8	
SO_2	0	–94	
CFC-11	4,800	3,300	4,600
CFC-12	11,000	6,100	10,600
CFC-113	6,200	4,700	6,000
CFC-114	10,000	9,800	9,800
CFC-115	7,500	7,400	7,200
CCl_4	1,800	1,300	1,800
MCF (CH_3CCl_3)	2,100	2,000	140
HCFC-22	2,500	2,500	1700
HCFC-123	150	150	120
HCFC-124	700	700	620
HCFC-141b	880	870	700
HCFC-142b	3,000	3,000	2,400
HCFC-225	700	700	180
Halon-1211	1,300	–12,000	1,300
Halon-1301	7,200	–62,000	6,900
CH_3Br	3	–824	5
HFC-23	13,100	13,100	12,000
HFC-32	1,000 [a]	1,000 [a]	550
HFC-43-10	5,800	5,800	1,500
HFC-125	5,300	5,300	3,400
HFC-134a	1,900	1,900	1,300
HFC-143a	6,200 [a]	6,200 [a]	4,300
HFC-227ea	4,900	4,900	3,500
HFC-245ca	970	970	640
CF_4	6,100	6,100	5,700
C_2F_6	14,700	14,700	11,900
SF_6	23,400	23,400	22,200

[a] For HFC32 and HFC143 and HFC227 the GWP of 10% is taken because the values for 1% slightly differ due to rounding errors.

of CH_4, CO, NO_x and NMVOC, while in the calculation of IPCC (2001b) constant atmospheric OH concentration was assumed. With the assumption that the emissions of CH_4, CO, NO_x and NMVOC will increase in the next century (see Fig. 4) the calculated lifetimes of IMAGE (see Eq. 2) will be higher resulting in a higher GWP.

2.6 Normalisation

In order to show the relevancy of our results, we caclulated sub-specific and total normalisation factors in CO_2 equivalents for global warming, base on greenhouse gas emission data for the world in year 2000. Emission data were taken from Wegener Sleeswijk et al. (2007). The results are given in **Table 5**.

Table 5: Total CO_2 equivalents (kg) for the year 2000 calculated with GWPs from Table 3 for direct and total effects

	Emission 2000 (kg)	CO_2 eq. direct (kg)	CO_2 eq. total (kg)
CO_2	$2.85\ 10^{13}$	$2.85\ 10^{13}$	$2.85\ 10^{13}$
N_2O	$1.15\ 10^{10}$	$3.16\ 10^{12}$	$3.16\ 10^{12}$
CH_4	$2.99\ 10^{11}$	$5.38\ 10^{12}$	$8.37\ 10^{12}$
CO	$1.04\ 10^{12}$	0	$3.13\ 10^{12}$
NO_x	$1.16\ 10^{11}$	0	$-1.16\ 10^{11}$
NMVOC	$1.67\ 10^{11}$	0	$1.33\ 10^{12}$
SO_2	$1.30\ 10^{11}$	0	$-1.22\ 10^{13}$
CFC-11	$4.06\ 10^{07}$	$1.95\ 10^{11}$	$1.34\ 10^{11}$
CFC-12	$1.01\ 10^{08}$	$1.11\ 10^{12}$	$6.14\ 10^{11}$
CFC-113	$3.86\ 10^{06}$	$2.39\ 10^{10}$	$1.81\ 10^{10}$
CFC-114	$2.07\ 10^{06}$	$2.07\ 10^{10}$	$2.03\ 10^{10}$
CFC-15	$8.73\ 10^{05}$	$6.54\ 10^{09}$	$6.46\ 10^{09}$
CCl_4	$4.17\ 10^{05}$	$7.51\ 10^{08}$	$5.42\ 10^{08}$
MCF (CH_3CCl_3)	$3.57\ 10^{05}$	$7.49\ 10^{08}$	$7.13\ 10^{08}$
HCFC-22	$3.00\ 10^{08}$	$7.51\ 10^{11}$	$7.51\ 10^{11}$
HCFC-124	$3.93\ 10^{06}$	$2.75\ 10^{09}$	$2.75\ 10^{09}$
HCFC-141b	$1.66\ 10^{08}$	$1.46\ 10^{11}$	$1.45\ 10^{11}$
HCFC-142b	$5.09\ 10^{07}$	$1.53\ 10^{11}$	$1.53\ 10^{11}$
Halon-1211	$4.82\ 10^{06}$	$6.26\ 10^{09}$	$-5.78\ 10^{10}$
Halon-1301	$9.26\ 10^{05}$	$6.67\ 10^{09}$	$-5.74\ 10^{10}$
HFC-125	$7.40\ 10^{06}$	$3.92\ 10^{10}$	$3.92\ 10^{10}$
HFC-134a	$1.30\ 10^{08}$	$2.47\ 10^{11}$	$2.47\ 10^{11}$
HFC-143a	$5.40\ 10^{06}$	$3.35\ 10^{10}$	$3.35\ 10^{10}$
SF_6	$5.22\ 10^{06}$	$1.22\ 10^{11}$	$1.22\ 10^{11}$
Total		$3.99\ 10^{13}$	$3.43\ 10^{13}$

Emissions are taken from Wegener Sleeswijk et al. (2007). GWPs for marginal emission changes of scenario A1 were used to obtain these data.

Assessing these results we see that the influence of indirect greenhouse gases, CO, NO_x, NMVOC and SO_2 is great. The impact of CH_4 becomes greater and Halons appear as a cooling factor, when indirect effects are taken into account. The latter will surely decrease in the coming century because of the expected decrease of these substances due to successful environmental policy (SRES 2000).

2.7 Uncertainties

Despite of the importance of indirect effects of greenhouse gases on the climate system, new accurate values of GWP cannot be determined yet due to uncertainties in these effects. **Fig. 5** shows the radiative forcings as IPCC reports them in its latest Assessment Report (IPCC 2007). Values of the well-mixed greenhouse gases are considered to be of high confidence on the level of scientific understanding (LOSU, IPCC 2007), whereas the indirect effects of aerosols are considered of low confidence (IPCC 2007). Also

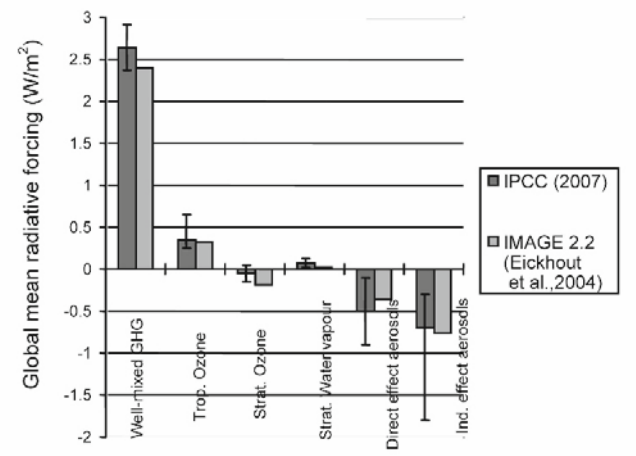

Fig. 5: Global and annual mean radiative forcing from 1750 to present according IPCC (2007) and according IMAGE 2.2 as presented in Eickhout et al. (2004). Only values that are reported in Eickhout et al. (2004) are taken from IPCC. IMAGE results are from 1765 to 1995; IPCC from pre-industrial to 2005

the broad uncertainty range that is attached to the indirect effect of aerosols clearly shows the current level of understanding. Forcings of aerosols are better understood than at the time of the Third Assessment Report (IPCC 2001) due to improved in situ, satellite and ground-based measurements and more comprehensive modelling, but remains the dominant uncertainty in radiative forcing (IPCC 2007). Therefore, this understanding needs to be improved further, before definite values of GWPs including indirect effects can be applied in LCA studies.

3 Conclusions

Our GWP values are generally equal with the values given in the IPCC report, but do in addition include indirect atmospheric effects such as the effects of changes in the ozone concentration and variable lifetimes. The inclusion of indirect effects in GWP values returns large differences in some important greenhouse gases and, therefore, is a valuable contribution to LCA-analyses. The simulations showed that the GWP values depend on the amount of emission change and the chosen background emission scenario. This can cause variations in the net GWP value up to a factor 2 for the amount of emission change and up to a factor 1.5 for background scenario. This variation is a point of interest for the use of GWPs. For LCA purposes where small emissions changes are considered the GWP for marginal contributions can be used. The user of the GWP can chose a background scenario, for instance A1 because it is most commonly used, but needs to be aware that the variation between different background scenarios is up to a factor 1.5, especially for CO_2.

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Supporting Information for Chapter 2

Table S1: Lifetimes and Mass-to-concentration factors for Greenhouse gases with constant lifetimes

Greenhouse gases with constant lifetimes	Mass to concentration factor cv (ppb/Tg)	Lifetime (yr); 1/k
CO_2	0.4688 [a]	[b]
N_2O	0.21	120
CFC-11	0.04314	45
CFC-12	0.04901	100
CFC-113	0.03163	85
CFC-114	0.03467	300
CFC-115	0.03836	1,700
CCl_4	0.03853	35
Halon-1211	0.03584	11
Halon-1301	0.03984	65
CH_3Br	0.0350877	0.7
CF_4	0.068	50,000
$C2F_6$	0.0508	10,000
SF_6	0.041	3,200

[a] The unit of the Mass concentration factor of CO_2 is ppmv/kg C.
[b] The lifetime of CO_2 is dependent on variable fluxes of CO_2 into the ocean and the biosphere.

Table S2: Reaction rates and Mass-to-concentration factors for Greenhouse gases with constant lifetimes

Greenhouse gas with non-constant lifetimes	Mass to concentration factor cv (ppb/Tg)	Reaction rate k_{GHG+OH} of the oxidation by OH (cm^3yr^{-1})	Transport losses to stratosphere and biosphere (yr^{-1})
CH_4	0.00040	$1.274 \cdot 10^{-7}$	18
MCF (CH_3CCl_3)	0.04689	$1.781 \cdot 10^{-7}$	2,100
HCFC-22	0.07234	$8.482 \cdot 10^{-8}$	2,500
HCFC-123	0.0409	$8.150 \cdot 10^{-7}$	150
HCFC-124	0.04583	$1.985 \cdot 10^{-7}$	700
HCFC-141b	0.05349	$1.144 \cdot 10^{-7}$	880
HCFC-142b	0.06224	$5.773 \cdot 10^{-8}$	3,000
HCFC-225	0.03082	$1.683 \cdot 10^{-7}$	700
HFC-23	0.086	$4.640 \cdot 10^{-9}$	13,100
HFC-32	0.116	$1.930 \cdot 10^{-7}$	900
HFC-43-10	0.07442	$9.236 \cdot 10^{-8}$	5,800
HFC-125	0.05211	$3.552 \cdot 10^{-8}$	5,300
HFC-134a	0.07442	$9.236 \cdot 10^{-8}$	1,900
HFC-143a	0.07142	$2.110 \cdot 10^{-8}$	6,100
HFC-227ea	0.035	$2.830 \cdot 10^{-8}$	4,900
HFC-245ca	0.0448	$1.660 \cdot 10^{-7}$	970

Table S3: Sensitivity coeffiecients for the formation of OH radicals and tropospheric ozone

(Indirect) Greenhouse gas	Sensitivity P in relation to the OH radical	Sensitivity R in relation to tropospheric ozone
CH_4	–0.32	6.7
CO	0.0042	0.17
NO_x	–0.000105	0.0014
NMVOC	–0.000315	0.0042

Table S5: Radiative efficiency coefficients for CO_2, CH_4 and N_2O and their initial pre-industrial concentrations in 1765

Green house gas	Radiative efficiency coefficient re	Pre-industrial concentration
CO_2	5.325 ($Wm^{-2}ppmv^{-1}$)	278 ppmv
N_2O	0.12 ($Wm^{-2}ppbv^{-1}$)	700 ppbv
CH_4	0.036 ($Wm^{-2}ppbv^{-1}$)	270 ppbv

The calculation of radiative forcing due to methane and nitrous oxide (equation 8 in the main article) is given by the equations I and II. The values for a and b are $2.01\ 10^{-5}$ and $5.31\ 10^{-15}$ respectively.

$$RF_{CH_4} = re_{CH_4}\sqrt{[CH_4]_t} - \sqrt{[CH_4]_{t0}} - \left\langle 0.47\ln\left[1 + a([CH_4]_t[N_2O]_{t0})^{0.75} + b[CH_4]_t([CH_4]_t[N_2O]_{t0})^{1.52}\right] - 0.47\ln\left[1 + a([CH_4]_{t0}[N_2O]_{t0})^{0.75} + b[CH_4]_{t0}([CH_4]_{t0}[N_2O]_{t0})^{1.52}\right]\right\rangle \quad (1)$$

$$RF_{CH_4} = re_{CH_4}\sqrt{[N_2O]_t} - \sqrt{[N_2O]_{t0}} - \left\langle 0.47\ln\left[1 + a([CH_4]_{t0}[N_2O]_t)^{0.75} + b[CH_4]_{t0}([CH_4]_{t0}[N_2O]_t)^{1.52}\right] - 0.47\ln\left[1 + a([CH_4]_{t0}[N_2O]_{t0})^{0.75} + b[CH_4]_{t0}([CH_4]_{t0}[N_2O]_{t0})^{1.52}\right]\right\rangle \quad (2)$$

Table S4: Radiative efficiency coefficients for halocarbons, SF_6 CO_2, CH_4 and N_2O and the number of chlorine and bromine atoms per compound

Green house gas	Radiative efficiency coefficient ($Wm^{-2}ppbv^{-1}$)	Number of chlorine or bromine atoms per compound
Trop. O_3	0.042 [a]	
CFC-11	0.25	3
CFC-12	0.32	2
CFC-113	0.3	3
CFC-114	0.31	2
CFC-115	0.18	1
CCl_4	0.13	4
MCF (CH_3CCl_3)	0.06	3
HCFC-22	0.2	1
HCFC-123	0.2	2
HCFC-124	0.22	1
HCFC-141b	0.14	2
HCFC-142b	0.2	1
HCFC-225	0.27	2
Halon-1211	0.3	1
Halon-1301	0.32	1
CH_3Br	0.01	1
HFC-23	0.16	0
HFC-32	0.09	0
HFC-43-10	0.4	0
HFC-125	0.23	0
HFC-134a	0.15	0
HFC-143a	0.13	0
HFC-227ea	0.3	0
HFC-245ca	0.23	0
CF_4	0.08	0
$C2F_6$	0.26	0
SF_6	0.52	0

[a] The unit of radiative efficiency of tropospheric ozone is $W^{-2}DU^{-1}$ (Dobson unit) Dobson units are used to express the concentration of tropospheric ozone.

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Expanded Damage Function of Stratospheric Ozone Depletion to Cover Major Endpoints Regarding Life Cycle Impact Assessment

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Background. Stratospheric ozone depletion is one of the important environmental issues for LCIA. The National LCA Project of Japan has developed a framework of LCIA since 1998, which tackles the issue employing an endpoint approach. Although the basic components were available in 2000, it was required that the target endpoints should be expanded in particular.

Objective. This study aimed at expanding the scope of damage function of ozone depletion in the LCIA framework of LIME. Damage function gives potential and quantitative damage for each endpoint per unit emission of ODS.

Methods. Marginal damage due to the unit emission of ODS was calculated for 13 substances for which quantitative information was available as follows: (1) the increase of UVB radiation at the earth's surface per unit emission of ODS was estimated, (2) the increase of potential damage per unit increase of UVB radiation was estimated, (3) the increase of potential damage per unit emission of ODS was determined by connecting the two relationships, and (4) correcting by the atmospheric lifetime of ODS, so that the damage function was then obtained. For other ODSs regulated by the Montreal Protocol, their damage functions were estimated by multiplying the ratio of ODP compared to the corresponding reference substance by the damage function of this reference substance.

Results and Discussion. The damage function of ozone depletion included the following endpoints: skin cancer and cataract for human health, crop production and timber production for social assets, and terrestrial NPP and aquatic NPP for primary productivity. And damage factors for each safeguard subject were also obtained.

Conclusion. The damage function of ozone depletion could cover all ODSs regulated by the Montreal Protocol and also cover important endpoints. Uncertainty of damage function is also an important point to be elucidated. Preliminary studies of uncertainty analysis have begun for the damage function of ozone depletion. However, further analysis is required to comprehensively evaluate the uncertainty of the damage function.